

SUPPORT FOR AMENDMENTS

Support for the amendments to the claims and the newly added claims can be found in the original claims & in the specification.

- Claim 1: original claim 8; page 7, lines 9-11 & page 8, lines 15-19;
- Claim 13: page 7, lines 9-11, 14-22 & page 8, lines 15-19;¹
- Claims 33-34: page 11, lines 5-8;
- Claim 35: 10, lines 10-11;
- Claim 36: Examples 1 & 5;
- Claim 37: page 10, lines 13-14 & the Examples; &
- Claims 38-39: first full paragraph on page 10.

No new matter has been added.

REMARKS/ARGUMENTS

The presently claimed anode materials are constructed so that the graphite powder thereof have interior portions of fine pores coated with a carbonized material of thermoplastic resin. There is no disclosure thereof in *Yoon*. In the remarks that follow, reference will be made to the publication of the present application, PG Pub US 2005/0158550 ("US '550"), rather than the specification as filed itself.

Included herewith is a Supplemental Declaration where Table 1 is, *inter alia*, amended to correctly recite the d(002) value for the natural graphite having 0 PVA (polyvinyl alcohol) added: 0.3354, rather than 0.3554 as in the Declaration filed April 16, 2009. This amendment corrects a typographical error.

Rejections under 35 U.S.C. § 102/103

1. The rejection of claims 1, 5, 8-12, 21, 29 & 31 under 35 U.S.C. § 102(e) or alternatively under 35 U.S.C. § 103(a) in view of U.S. Patent 6,596,437 ("*Yoon*") is respectfully traversed.

The Office alleges that *Yoon* discloses graphite powders coated with various carbonized materials of thermoplastic resins. The Office considers that characteristics of the presently claims would inherently be met by the *Yoon* graphite powders. The Office maintains that the alleged coating described in *Yoon* includes the scenario where the *Yoon* resins are coated within interior pores present in the *Yoon* graphite particles. *Yoon* controverts the Office's position.

Claims 1 and 13 have been amended to recite, in part, that the graphite powders display "...an average interlayer spacing d_{002} of not more than 0.3360 nm and ***L(112) of not less than 15 nm...***" There is no disclosure of L(112) values for the graphite powders of *Yoon*. This characteristic induces an increased discharge capacity in the resulting battery.¹ Furthermore, there is no disclosure in *Yoon* that the interlayer spacing d_{002} changes by no more than 0.0005 nm (claims 33 & 34), the importance thereof is discussed below. Moreover, the graphite powder of the present invention can be spherical in shape (see claim 35). *Yoon* discloses that the crystalline carbon (allegedly analogous to the graphite powder of the present invention) is "cylindrical graphite, plate graphite and fibrous graphite";² there is no disclosure of spherical-shaped graphite powder in *Yoon*. *Yoon* discloses that the amorphous carbon coats the outer surface of the crystalline carbon to form a shell around the crystalline carbon (see Abstract), rather than that the interior portions of fine pores within the graphite powders are coated.

¹ See [0027] of US '550.

² Col. 4:22-26 of *Yoon*.

The problem that the present invention attempts to solve is to use a graphite-type material as the active material for the anode while preventing decomposition of the propylene carbonate electrolyte. This decomposition is a known problem and occurs when the electrolyte contacts a graphite anode:

It is found that graphitized materials, including natural graphite, have the discharge capacity close to a theoretical value, while on the other hand, they have generally a large irreversible capacity of not less than a few tens of mAh/g resulting from the decomposition of electrolytic solution on the anode in the initial stage of electric charge. This is a major hurdle to realize a high performance lithium ion secondary battery. Particularly when propylene carbonate is used for the electrolytic solution, ***the electrolytic solution is significantly decomposed on the anode***. Due to this, the use of propylene carbonate for the electrolytic solution is largely restricted.³

Yoon, on the other hand, addresses this problem:

If propylene carbonate, which is less costly than ethylene carbonate and remains a liquid at room temperature, is applied as electrolyte in the battery using graphite as the negative electrode, graphite layers are peeled by co-intercalation of the electrolyte, and, as a result, lithium-ion intercalation is not properly realized. Accordingly, the capacity of the battery is reduced.⁴

The solution for *Yoon* is to chemically bond amorphous carbon to crystalline carbon (e.g. graphite):

Since the surface of the inventive negative electrode active material is either amorphous carbon or turbostratic structure carbon, it is possible to use polypropylene carbonate electrolyte.⁵

Thus, the surface of the *Yoon* active material is amorphous carbon, not crystalline carbon, and this surface contacts the electrolyte and allows for the use of polypropylene electrolyte. Thus, *Yoon* clearly describes a outer shell of amorphous carbon that is present on an outer surface of crystalline carbon. There is no disclosure that the amorphous carbon is introduced

³ [0002] of US '550, in part, emphasis added.

⁴ Col. 1:50-57 of *Yoon*.

⁵ Col. 6:24-27 of *Yoon*.

into fine pores within the crystalline carbon. On the other hand, the graphite powders of the present claims, when present in an anode which is present in a battery, are contacted by the propylene carbonate-comprising electrolyte (see claim 36).

Yoon discloses that the chemical bonding is accomplished by 1) functionalizing crystalline carbon to form $-OH$ groups on the outer surface of the crystalline carbon, and 2) reacting the $-OH$ groups present in the surface of the crystalline carbon with various functionalized resins so that the resins are chemically bonded to the surface of the crystalline carbon:

In the first process of the inventive second method above, it is preferable that the crystalline carbon is selected from the group consisting of cylindrical graphite, plate graphite and fibrous graphite. For the reactive functional group, it is possible to use any functional group which is able to chemically combine with amorphous carbon when introduced on a surface of crystalline carbon, preferably a hydroxyl group. A method of introducing a hydroxyl group on the crystalline carbon surface will now be described.

Crystalline carbon is contacted with sulfuric acid, preferably concentrated sulfuric acid. By processing the sulfuric acid, sulfate ($-HSO_4$) and hydrogen ($-H$) is added to a double bonding of the crystalline carbon surface. After sulfate ($-HSO_4$) is introduced on the double bonding of the crystalline carbon surface, the crystalline carbon is washed with water to substitute the sulfate ($-HSO_4$) with a hydroxyl group ($-OH$), thereby introducing the hydroxyl group.

In the second process of the inventive second method, the reactive functional group introduced on the surface of the crystalline carbon surface is chemically combined preferably with a hydroxyl group and an amorphous carbon precursor. It is preferable that the amorphous carbon precursor is a hard carbon precursor or a soft carbon precursor.

In the above, it is preferable that the hard carbon precursor is selected from the group consisting of polyimide resin, furan resin, phenol resin, polyvinyl alcohol resin, cellulose resin, epoxy resin and polystyrene resin. The hard carbon precursor includes a hydroxyl group and other functional groups which can react with the hydroxyl group introduced on the surface of the crystalline carbon. Accordingly, the functional group of the hard carbon precursor condensation-reacts with the hydroxyl

group introduced on the surface of the crystalline carbon to realize an ether combination, thereby producing a combination material of the crystalline carbon and the hard carbon precursor.⁶

Thus, as *Yoon* clearly states, material allegedly analogous to the "carbonized materials of thermoplastic resins" of the present claims is chemically bonded to the surface of the crystalline carbon, thereby forming an "amorphous carbon or turbostratic structure ***carbon shell around*** the crystalline carbon."⁷ Thus, it would be expected that the interlayer spacing between the crystalline carbon core and the carbon shell would be different if the carbon shell is on the surface of the crystalline carbon core rather than within the crystalline carbon core.

The *Yoon* graphite materials are analyzed by X-ray diffraction and argon ion laser Raman spectroscopy, which clearly show a difference between the crystalline carbon core and the carbon shell.

Using X-ray diffraction and argon ion laser Raman spectroscopy to measure characteristics of the core of the negative electrode active material, it was discovered that a distance (d_{002}) between layers of the crystalline carbon is preferably 3.35-3.42 Å, and that a ratio (I_{1360}/I_{1580}), in an argon ion laser Raman spectra, between a peak value at 1360 cm^{-1} to a peak value at 1580 cm^{-1} is preferably 0.05-0.4.

Further, using X-ray diffraction and argon ion laser Raman spectroscopy to measure characteristics of the core of the negative electrode active material, it was discovered that a distance (d_{002}) between layers of the amorphous carbon or turbostratic structure carbon shell is preferably 3.38-3.80 Å, and more preferably 3.40-3.700 Å. In addition, in an argon ion laser Raman spectra, it is preferable that a ratio (I_{1360}/I_{1580}) between a peak value at 1360 cm^{-1} to a peak value at 1580 cm^{-1} is 0.3-1.5.⁸

Here, two distinct ranges in the d_{002} values determined by X-ray diffraction are observed between the crystalline carbon core (3.35-3.42 Å) and the X-ray diffraction pattern of the amorphous carbon or turbostratic structure carbon shell (3.38-3.80 Å). Thus, the minimum

⁶ Col. 4:22-58 of *Yoon*, emphasis added.

⁷ See Col.5:21-22 of *Yoon*.

⁸ Col. 5:36-42 & Col. 5:57-65 of *Yoon*.

value for the graphite particles upon treatment is 3.38 Å (0.3380 nm). On the other hand, the minimum value for the coated graphite particles of the present invention is 0.3360 nm (see claims 38-39).

Likewise, the ratio between the intensities of the peaks measured at 1360 cm⁻¹ and 1580 cm⁻¹ shifts between the crystalline carbon (0.05-0.4) and the amorphous carbon (0.3-1.5). If the amorphous carbon allegedly analogous to the resins of the present claims was located only in the interior fine pores of the crystalline carbon as alleged by the Office, then there should be no such shift in the values for these characteristics between pre- and post-treatment with the thermoplastic resin. Moreover, the enclosed Supplemental Declaration supports the shift in d_{002} in the *Yoon* graphite powders:

5. Graphite powder was also prepared by the process of Example 1 disclosed in U.S. Patent 6,596,437. The average interlayer spacing $d(002)$ of the graphite powder without a resin coating was determined by X-ray diffraction: $d(002) = 3.35 \text{ Å}$. The average interlayer spacing $d(002)$ was then determined for the graphite powder coated with a resin: $d(002) = 3.41 \text{ Å}$. Thus, a substantial increase in the interlayer spacing of the graphite powder prepared by the process of Example 1 in U.S. Patent 6,596,437 is observed.

Thus, the change in d_{002} observed when the *Yoon* experiment was repeated is 0.006 nm. On the other hand, the change in the X-ray pattern for the presently claimed anode materials is preferably at most 0.0005 nm, a full order of magnitude less than what is observed for *Yoon*:

This coating can keep the diffraction line in the X-ray diffraction substantially unchanged. When an increasing amount of average interlayer spacing d_{002} defined by the Gakushin-method for X-ray diffraction of carbon is 0.0005 nm or less, the capacity of the graphite powder used as a core can be used effectively.⁹

See claims 33-34. This distinguishing characteristic of the present claims can be seen in the enclosed Supplemental Declaration. In Table 1 thereof

⁹ [0028] of US '550.

Table 1: Average interlayer spacing d(002) of anode material before and after resin coating.

Added amount of PVA (parts by weight with respect to 100 parts by weight of natural graphite)	d(002) (nm)
0 (no coating)	0.3354
10	0.3354
50	0.3355
100	0.3354

*Although 50 parts by weight of PVA is adopted in the examples of the present application, it is understood that d(002) does not change even in the case of 100 parts by weight.

the d_{002} values for graphite powders with 0, 10, 50, and 100 parts of PVA (relative to 100 parts by weight of natural graphite) is reported. Notice that there is no change when 10 and 100 parts of PVA are added, while a difference of 0.0001 nm is observed for 50 parts. Thus, Applicants have clearly demonstrated that interior portions of fine pores within the graphite powders are coated in the presently claimed anode materials, while, on the other hand, the crystalline carbon of *Yoon* is coated on the exterior.

The Office dismissed the Declaration filed April 16, 2009 by stating: "Examiner is not convinced with the logical assumption that a showing of the difference in average interlayer spacing equates to the presence or not of resin coated interior portions of the fine pores. Applicant's specification does not correlate interlayer spacing with the presence of resin coated interior portions of the fine pores, and neither does the aforesaid Declaration."

Contrary to the Office's statement, US '550 states at [0022] and [0028]:

The term, "coating" here in the present invention means the coating of interior portions of the fine pores, rather than the coating of surfaces of the particles.

This coating can keep the diffraction line in the X-ray diffraction substantially unchanged. When an increasing amount of average interlayer spacing d_{002} defined by the Gakushin-method for X-ray diffraction of carbon is 0.0005 nm or less, the capacity of the graphite powder used as a core can be used effectively.

Thus, the change in interlayer spacing is correlated to the coating of interior portions of fine pores in Applicant's specification. Moreover, a description of the correlation between d_{002}

value and the coating of pores can be found in paragraphs 7-11 of the enclosed Supplemental Declaration.

The Office further dismissed the Declaration by stating: "At the outset it is unclear as to the type of phenol resin used in the experimentation of Example 1, especially given that polyvinyl alcohol is one of the claimed limitations. Even largely assuming that polyvinyl alcohol was used, Examiner is not convinced with the logical assumption that a showing of the difference in average interlayer spacing equates to the presence or not of resin coated interior portions of the fine pores." Example 1 of *Yoon* states that "phenol resin" was used. Applicants reproduced this experiment, which is what is required per MPEP § 716.02(e): "An affidavit or declaration under 37 CFR 1.132 must compare the claimed subject matter with the closest prior art to be effective to rebut a prima facie case of obviousness." Thus, Applicants have compared the claimed subject matter with that which is disclosed in *Yoon*.

Additional support for the difference between the *Yoon* coating on the crystalline carbon and the coatings of the present claims can be found in the methods of making the respective powders. In *Yoon*, the crystalline carbon is first treated with sulfuric acid to introduce -OH groups on the surface of the crystalline carbon. These groups are used to chemically bond the amorphous carbon to the crystalline carbon. See quote from Col. 4 of *Yoon*, above. On the other hand, the methods for making the presently claimed anode materials involve dry-blending and baking the graphite powder and the thermoplastic resin *in the presence of nitrogen or argon gas* (see claim 21). These gases are inert, and are usually introduced to ensure that chemical reactions are inhibited. Thus, one of ordinary skill in the art would readily see an important difference between the processes of *Yoon* and the processes of the present claims.

The characteristics (1) to (6) are not inherent to the alleged analogous graphite powders of Yoon. MPEP § 2163.07(a) states in part: "To establish inherency, the extrinsic evidence 'must make clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill.'" These features are necessary for producing batteries having improved irreversible capacities. Since *Yoon* forms a carbon shell around the graphite particles rather than mainly filling fine pores in the graphite particles, none of these characteristics would be inherent to the particles of *Yoon*.

For all of the reasons given above, *Yoon* neither anticipates nor renders obvious the anode materials of the present claims. Thus, one of ordinary skill in the art would not be led to coat interior portions of fine pores present within graphite powders based on the disclosure of *Yoon*.

Accordingly, the rejection is no longer tenable and should be withdrawn.

2. The rejection of claims under 35 U.S.C. § 13-20, 28, 30 & 32 103(a) in view of *Yoon* and JP 2001-196097 ("*Aihara*") is respectfully traversed for the same reasons given above. *Aihara* contains no teaching of the dry-blending and baking of the present claims, nor does it contain any disclosure of coating interior portions of fine pores of graphite powders with thermoplastic resins versus coating the outer portion of crystalline carbon with a thermoplastic resin. Accordingly, the rejection is no longer tenable and should be withdrawn.

Conclusion

Applicants respectfully submit that the above-identified application is in condition for allowance. Notification thereof is requested.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
MAIER & NEUSTADT, L.L.P.



Benjamin A. Vastine, Ph.D.
Registration No. 64,422

Customer Number

22850

Tel: (703) 413-3000
Fax: (703) 413 -2220
(OSMMN 07/09)